Appendix 2B-1: The Effect of Dryout and Burn on the Everglades Mercury Cycle

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EXECUTIVE SUMMARY

Following an extended period of no rain, some areas of the Everglades dried out completely, and a subset of those areas burned. An *ad hoc* study was organized to evaluate the effect of this infrequent but not uncharacteristic phenomenon on the Everglades mercury cycle. The study was co-funded and carried out by the USGS District Office in Middleton, WI, and the South Florida Water Management District in West Palm Beach, FL. Samples of surface water, pore water, soil, periphyton, and mosquitofish were collected at ten sites in July 1999 about six weeks after re-inundation. Three of these sites, two of which had burned, had never been sampled before, while the remaining sites had been routinely sampled in the period 1995-1999 as part of the USGS Aquatic Mercury Cycling in the Everglades (ACME) study. Follow-up sampling by District staff collected all but pore water at a select subset of the study sites in August, October, and November 1999. The study results demonstrate that the effect of severe dryout on the Everglades mercury cycle is profound, with concentrations of methylmercury increasing in soil pore water 5- to 35-fold over historical average levels within six weeks of reinundation. These pulses were followed by substantial increases in the concentrations of methylmercury in periphyton mats and mosquitofish, peaking within 60-90 and 90-120 days of reinundation, respectively. There is also some evidence that these methylmercury pulses continued up the Everglades food chain into top-predator fish (i.e., largemouth bass) in the fall of 1999, persisting into the fall of 2000, and in the eggs of top-predator wading birds (i.e., the great egret) in the spring of 2000, persisting into the spring 2001. The most recent data suggest that this pulse is clearing from the Everglades ecosystem, however. An analysis of surface water, pore water, and soils data collected along a well-studied nutrient gradient in the northern Everglades indicates that readily oxidized species like iron and sulfur also exhibited pulsed increases in pore water concentrations where dryout occurred, but that these responses did not persist, returning to historical average levels at all but one site within three quarterly sampling events of rewetting. No such "first-flush" effect on pore water phosphorus was observed. Proper interpretation of mercury temporal trends in Everglades water, sediment, and biota requires cognizance of the timing and severity of such events. Moreover, changes in Everglades hydrology that decrease the magnitude, spatial extent, or frequency or recurrence of severe dryout events could have a beneficial effect on the Everglades mercury problem.

INTRODUCTION

This is a follow-up to an earlier publication on the same topic (Krabbenhoft and Fink, 2001). It provides a more detailed conceptual model of the effect of dryout and rewetting on the mercury cycle and updates the figures depicting the effect of the 1999 dryout on surface water, soil, and pore water chemistries along a well-studied nutrient gradient in Water Conservation Area 2A (WCA-2A) in the remnant northern Everglades.

BACKGROUND

A severe dryout of the northern Everglades occurred in the winter and spring of 1999, with several areas experiencing fires ranging in severity from plant tops only to deep peat burns (ECR, 2001). The dried and burned areas are depicted in Figure 1. The dryout interrupted routine monthly monitoring of surface water along the WCA-2A nutrient gradient ("F" Transect; Figure 2) in February through May 1999 because there was no water to sample at several of the "F" Transect sites and/or airboat access was precluded. However, quarterly monitoring of soil (0-5 cm) and pore water (10-20 cm) was not interrupted. The "F" Transect is comprised of Sites F1, F2, F3, F4, F5, and U3, ranging from an average water column total phosphorus concentration from about 70 ppb at F1 (1.8 km downstream of S-10C) to about 8 ppb at U3 (10,8 km downstream of S-10C), with a steep, exponentially decreasing concentration gradient in between (McCormick et al., 1999).

CONCEPTUAL MODEL

One of the most significant influences on MeHg production and subsequent bioaccumulation is the drying and rewetting cycle of the Everglades. During the drawdown period preceding dryout, soil pore water chemistry and the rate of exchange of pore water constituents with the overlying water have been observed to change (Reddy et al., 1999). As soil dryout proceeds, it can be confidently predicted that carbon, sulfur, and iron species in surficial soils are oxidized, albeit to different degrees and at different rates (Dmytriw et al., 1995; Yin et al., 1997; Gun et al., 2000; Taillfert et al., 2000; W. Orem, USGS, personal communication, 2000; Fink, 2001). Oxidized iron species have been demonstrated to sorb inorganic mercury, Hg(II) (Lockwood and Chen, 1974). Reinundation of oxidized soils is usually accompanied by a "first-flush" release of nutrients (Newman and Pietro, 2001), iron (Krabbenhoft and Fink, 2001), and trace metals, including inorganic mercury (Regnell, 1994; Dmytrw et al., 1995), from the inorganic and organic binding sites in the soil. There is also evidence of a first flush of sulfate (Krabbenhoft et al., 2000; Krabbenhoft and Fink, 2001; Fink, 2001). Following the first-flush release of sulfate and the onset of conditions devoid of dissolved oxygen (anoxic or anaerobic conditions), the metabolic activity of sulfate-reducing bacteria is likely to be stimulated, with an associated stimulation of methylmercury production (Gilmour et al, 1992). Thereafter, sulfide produced by sulfate-reducing bacteria begins to accumulate in the pore water of sediment or hydrated soil (Krabbenhoft et al., 2000; Krabbenhoft and Fink, 2001; Fink, 2001). The reduced sulfur in soil organic matter has a high affinity for Hg(II) (Xia et al., 1999). In addition, under some conditions of sulfate loading and eutrophication, pore water sulfide can build up to potentially phytotoxic levels (Lamers et al., 1998).

During this period, it has been hypothesized that the presence of high concentrations of short-chain carboxylic acids, sulfate, and inorganic mercury in readily bioavailable forms accelerate methylmercury production until they are reduced and/or sequestered into less bioavailable forms by biotic or abiotic processes (Krabbenhoft et al., 2000; Krabbenhoft and Fink, 2001). The inferred presence of excess dissolved iron following rewetting (Krabbenhoft and Fink, 2001) may also contribute to this pulsed production (Howard, 1993; Marvin-DiPasquale et al., 2001). The excess methylmercury produced in this fashion will then follow the transport and fate pathways outlined above, resulting in a net transfer of excess methylmercury directly or indirectly into the Everglades aquatic food chain.

If the duration of accelerated methylmercury production is short, because the soil pools of labile, bioavailable sulfate, carbon, and inorganic mercury are small and rapidly consumed, then the total mass of methylmercury produced will be small and the magnitude and duration of subsequent excessive bioaccumulation of methylmercury in top-predator fish and their predators will be short-lived. This is the so-called “first flush effect.” Conversely, if these pools are large or there is an external source of the limiting factor capable of sustaining a high, first-flush methylmercury production rate for a long time, then the first-flush mass of methylmercury produced will be large. It will then result in excessive bioaccumulation at the top of the food chain, and it will clear more slowly from the ecosystem than a “first-flush” pulse, because (1) top-predator fish eliminate and dilute methylmercury through growth only slowly (Norstrom et al, 1976; Rodgers, 1994) and are long-lived; and (2) the methylmercury present in the food web is more efficiently recycled back into the food web than methylmercury in water, sediment, or sorbed to particles. This results in the so-called “reservoir effect,” first observed in hydroelectric reservoirs created by flooding forested glacial till soils in northern temperate regions (Bodaly et al, 1984; Scruton et al., 1994; Rodgers et al., 1995) but also observed in natural, created, or expanded wetlands (St. Louis et al., 1994; St. Louis et al., 1996; Kelly et al., 1997; Paterson et al., 1998). This has also resulted in the increase in methylmercury body burdens in insect-eating birds (Gerrard and St. Louis, 2001) and fish-eating birds and mammals foraging in these water bodies (Wolfe et al., 1994).

However, if labile, bioavailable sulfate is present in substantial excess, surficial sediments remain anaerobic, and no other factor limits microbial metabolism or affects sulfur speciation, then sulfide can accumulate to concentrations that actually inhibit methylmercury production (Craig and Bartlett, 1978; Compeau and Bartha, 1985; Berman and Bartha, 1986; Chen et al., 1997; Gilmour et al., 1998b; Benoit, 1999a,b; Jay et al, 2000; Benoit et al., 2001; Marvin-DiPasquale et al., 2001). This may also explain why the constructed wetland, STA-1W, which was farmed and fertilized prior to being reclaimed as a constructed wetland, returned to baseline concentrations of methylmercury in water and mosquitofish within a few months of the onset of the “first-flush” effect (Rawlik, 2001a,b), while STA-2 Cell 1, which was never farmed, has not after two years of start-up efforts (Rumbold and Fink, 2002). It has been hypothesized with moderate confidence (Gilmour et al, 1998b) that sulfide inhibition is causing eutrophic Everglades regions with conditions otherwise deemed ideal for methylmercury production (e.g., ENR Project and WCA-2A-F1) to exhibit low methylmercury production and correspondingly low concentrations in fish at all trophic levels (Cleckner et al., 1998; Lange et al., 1998, 1999; Loftus et al., 1998; Rumbold et al., 2000; Rawlik, 2001a,b; Rumbold et al., 2001). Conversely, unimpacted or virtually pristine areas in the Everglades exhibit much higher methylmercury production rates (e.g., WCA-2A-U3 and WCA-3A-15) and correspondingly higher concentrations in fish at all trophic levels. This effect may be amplified by two- to seven-fold as a result of the addition of another step in the food chain (USEPA, 1997; Lange et al., 1998, 1999; Loftus et al., 1998) due to improved water quality (i.e, higher dissolved oxygen, lower pore water sulfide) or improved habitat (i.e., more open, deeper water). Based on an extensive, three-year study by USGS, both the fraction of methylmercury that is total mercury in surficial soils and the concentration of total mercury as a surrogate for methylmercury in mosquitofish are strongly inversely correlated with pore water sulfide in surficial soils across the Everglades (Fink, 2002).

STUDY SITE AND METHODS

The rains began at the end of May 1999, and District routine monitoring and special mercury sampling resumed in mid-June 1999. In response to this relatively infrequent but natural phenomenon, the U.S. Geological Survey-Middleton, Wisconsin, and the District collaborated in what became known as the Post-Burn Study. USGS staff collected samples at several of the sites monitored as part of its Aquatic Cycling of Mercury in the Everglades (ACME) Project in the period 1995-1999, as well as several new burned sites, beginning about six weeks after reflooding in July 1999. Water, pore water, soil, periphyton, and mosquitofish were collected at the sites depicted in Figure 3. In addition to total mercury and methylmercury, the surface water, pore water, and soil were analyzed for a suite of constituents known or reasonably anticipated to influence methylmercury production or bioaccumulation. District staff then continued to collect water, soil, periphyton, and mosquitofish at several of these sites in August, October, and November 1999 (Figure 3). USGS sampling protocols were followed by both field crews. All environmental samples were analyzed by USGS-Middleton according to published procedures (e.g., Krabbenhoft et al., 1998).

RESULTS AND DISCUSSION

The effect of severe dryout on soil pore water chemistry in the Everglades is illustrated in Figures 4 and 5, which depict the yearly average concentrations for “F” Transect pore water sulfide and iron, respectively, and Figure 6, which depicts the ratio of the yearly average pore water sulfide (ug/L) and sulfate (mg/L) concentrations, for reporting years 1995-1996 through 1999-2000. Note the 2- to 10-fold increase in the <u>average</u> filtered iron values, the 3- to 6-fold increase in the average filtered sulfate values, and the 2- to 7- increase in the average sulfide-to-sulfate ratio for 1999-2000 relative to the preceding four years. If there was a post-dryout response of pore water filtered phosphate, it is not evident in the annual averages (Figure 7). In the succeeding year, these parameters returned to more typical values, with the exception of F4, where pore water iron remained high and the sulfide-to-sulfate ratio remained low. The reason for this anomalous behavior is unknown. To better resolve the timing, magnitude, and duration of post-dryout changes in pore water chemistry, data for the quarters sampled in March, June, and October 1999 are illustrated in Figures 8, 9, 10, and 11 for filtered iron, filtered sulfate, filtered sulfide, and filtered phosphate, respectively. The extreme responsiveness of pore water iron, sulfate, and sulfide but not phosphate to the post-dryout reflooding of northern WCA-2A in late May 1999 is apparent in the June 1999 data. At the most and least eutrophic sites, F1 and U3, the pore water sulfide response peaked in June and March 1999, respectively, while the pore water sulfide peak at the intervening sites was not reached until October 1999.

Although chemical speciation was not carried out, based on the above discussion, it is likely that the reduced iron (Fe^{+2}) and sulfur ($S^{=}$), which are generally more strongly sorbed, complexed with organic matter, and precipitated than oxidized iron (Fe^{+3}) or sulfur ($SO_4^{=}$), were oxidized and liberated during the severe dryout of northern WCA-2A. It has been hypothesized that the liberation of excess sulfate then stimulated excess methylmercury production (Krabbenhoft et al., 2000; Krabbenhoft and Fink, 2001), as evidenced by the ratios of the concentrations of MeHg in soil (0-5 cm) to historical averages at Sites F1, U3, and 3A-15 in the period July through November 1999 (Figure 12). It has also been hypothesized that the observed rapid decrease in the excess methylmercury in soil was caused by the depletion of the excess pore water sulfate pulse (Krabbenhoft et al., 2000; Krabbenhoft and Fink, 2001). An alternative hypothesis (Fink, 2002) is that, as the sulfate was depleted, pore water sulfide accumulated to concentrations that inhibited excess methylmercury production, as has been inferred from laboratory experiments (Benoit et al., 1999; Jay 2000; Benoit et al., 2001a,b) and field observations (Gilmour et al., 1998a,b; Gilmour et al, 1999) as anaerobic conditions returned and the excess pore water sulfate was depleted.

The excess methylmercury production pulse subsequently manifested itself in mosquitofish at highly eutrophic site F1 almost immediately after reflooding, peaking in June 1999 at 3.5 times the average site value, while site U3 peaked about 90 days later at 3.5 times its average value, but Site 3A-15, which did not dry out, never significantly exceeded its average value during the wet season months (Figure 12). Subsequent upturns in total mercury concentrations in sunfish and largemouth bass were observed in largemouth bass collected from northern Everglades sites in the fall of 1999, and this response appears to have persisted through the fall of 2000. Thereafter, a downturn in fish mercury levels was observed in the fall of 2001 (Figure 13; Rumbold et al., 2002). This pattern was mimicked in wading bird eggs collected in the spring of 2000, 2001, and 2002 (Figure 14; Rumbold et al., 2002).

CONCLUSIONS

Based on the preceding, clearly an extensive, prolonged dryout has a substantial, potentially ecologically significant impact on mercury biogeochemistry and bioaccumulation in the northern and central Everglades. Thus, efforts to increase the hydroperiod and decrease the pyroperiod of the Everglades could have a beneficial effect on methylmercury bioaccumulation, all other factors being equal.

[To Be Supplied]

Figure 1. Everglades areas that dried out and burned in the spring of 1999

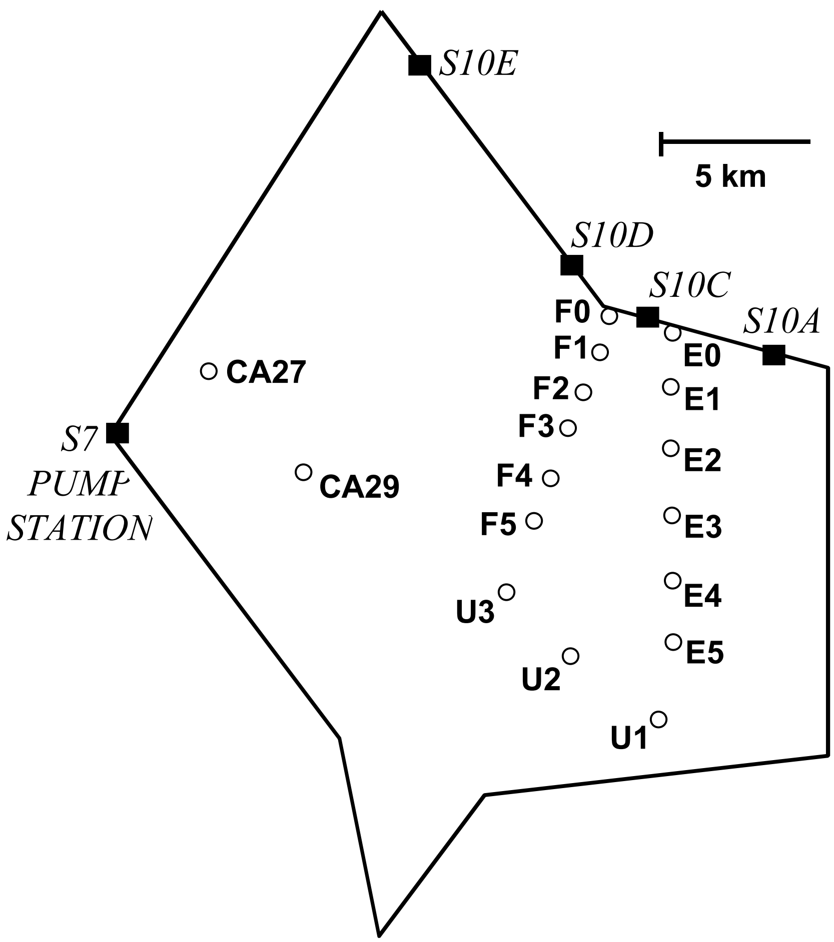

Figure 2. WCA-2A nutrient gradient study sites, focusing on the "F" transect

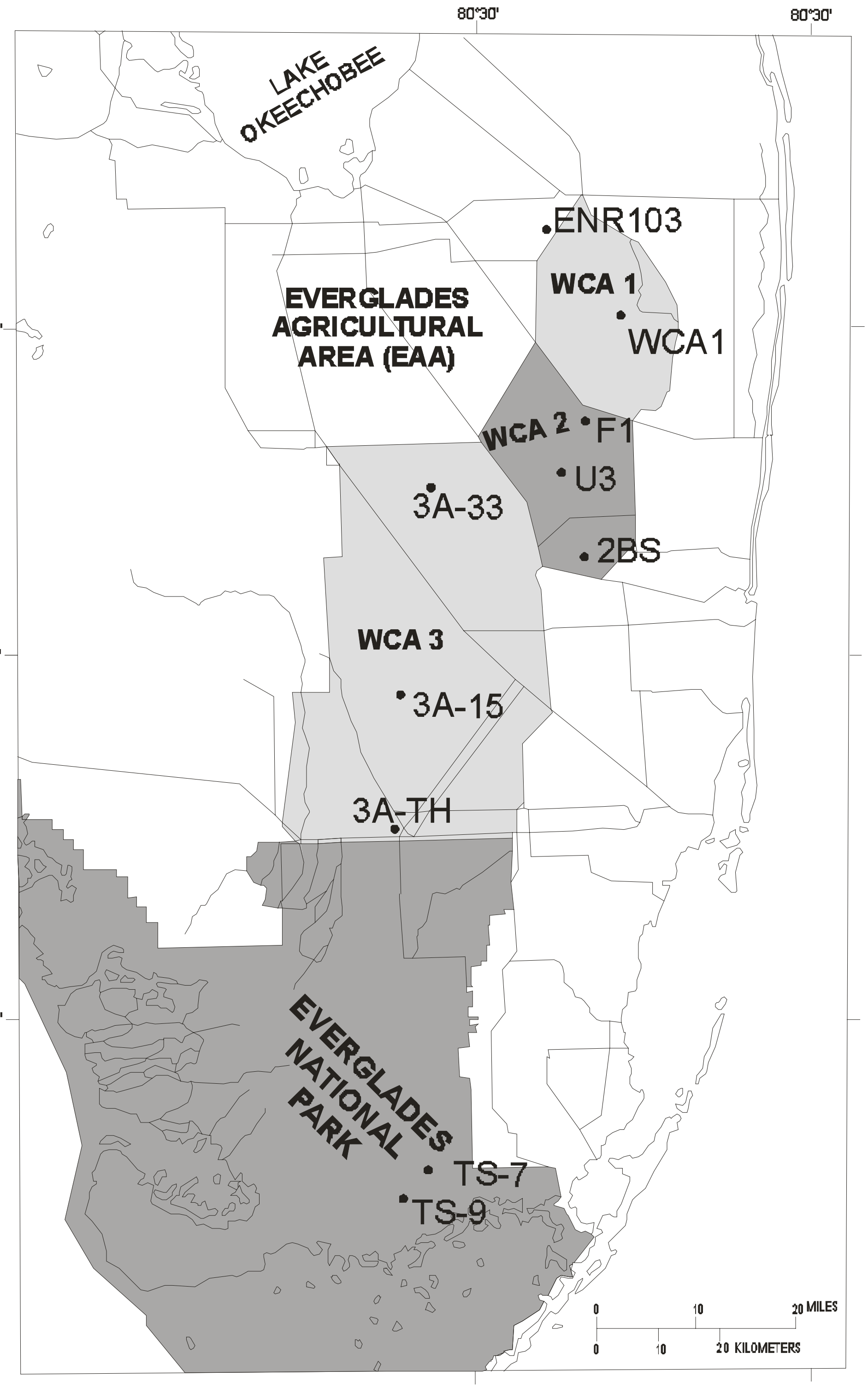

Figure 3. Post-burn study sites in the northern and central Everglades

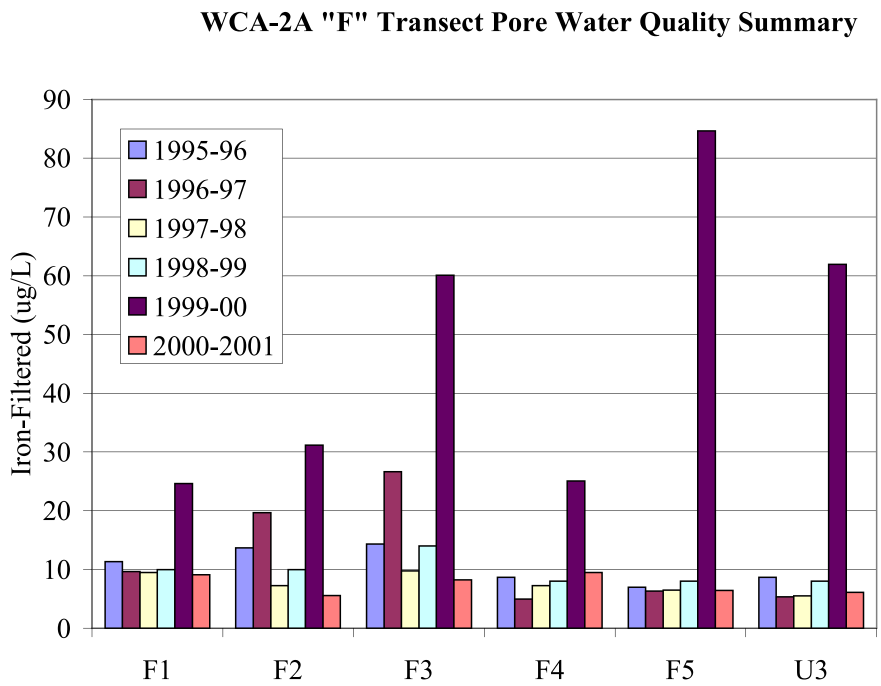

Figure 4. Reporting-year average pore water total iron concentrations (10-20 cm) for the period of record along the "F" transect in WCA-2A

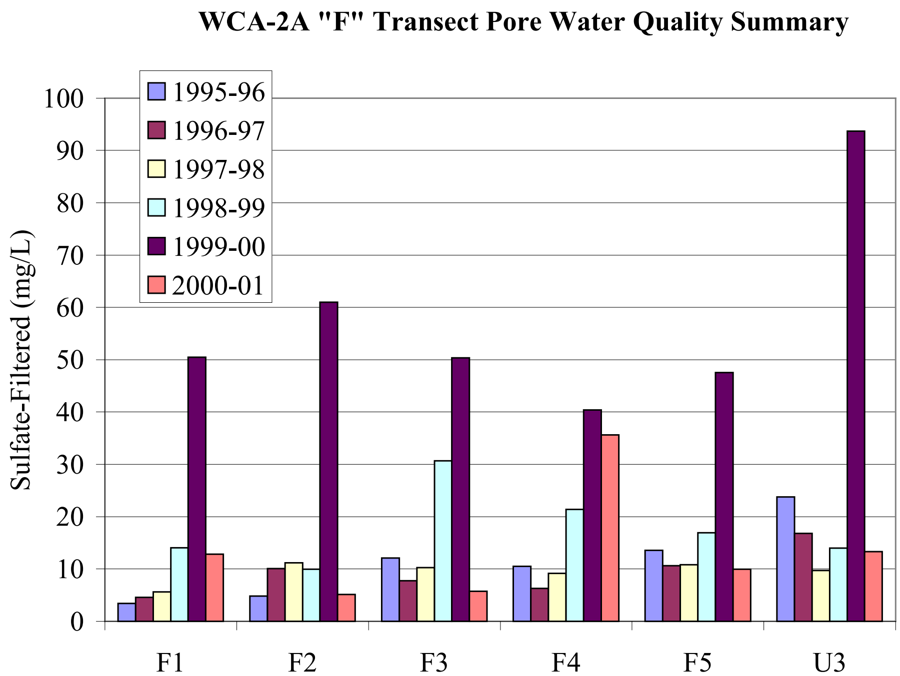

Figure 5. Reporting-year average pore water sulfate concentrations (10-20 cm) for the period of record along the "F" transect in WCA-2A

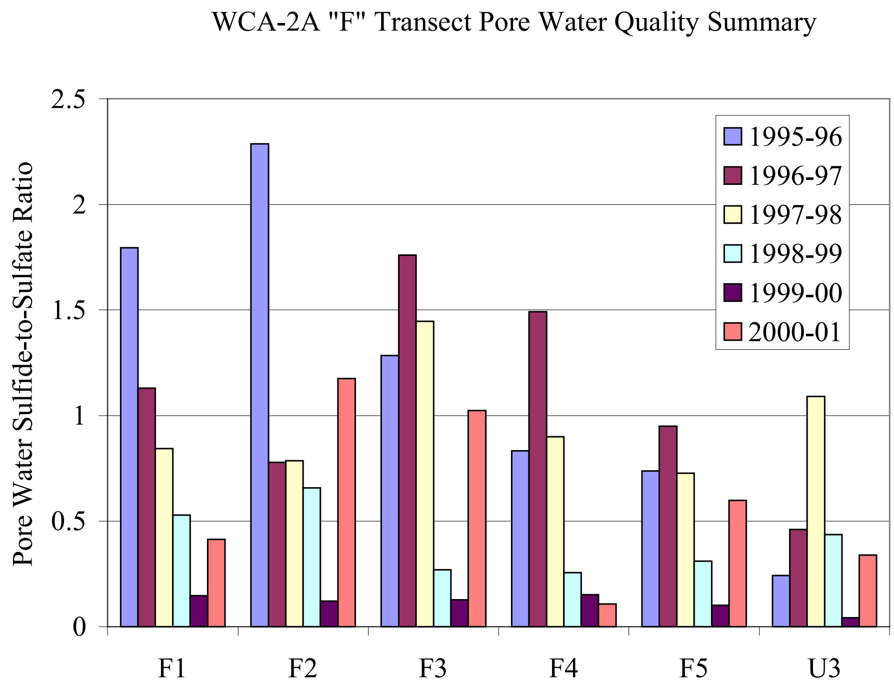

Figure 6. Reporting-year average ratio of pore water sulfide-to-sulfate concentrations (10-20 cm) for the period of record along the "F" Transect in WCA-2A. [Sulfide concentrations in ug/L; sulfate concentrations in mg/L)

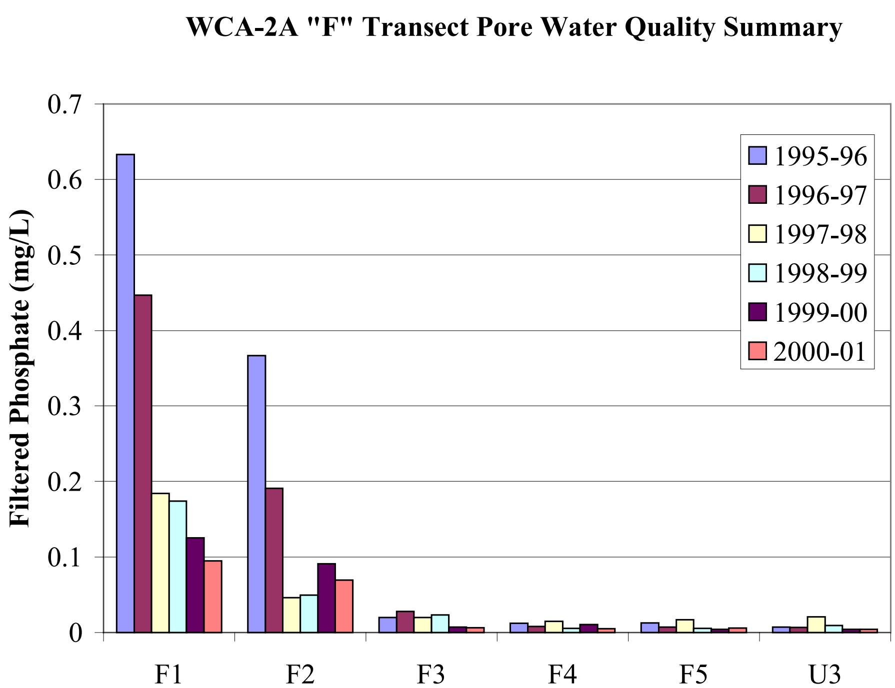

Figure 7. Reporting-year annual average pore water concentrations of filtered phosphate (10-20 cm) for the period of record along the "F" Transect in WCA-2A

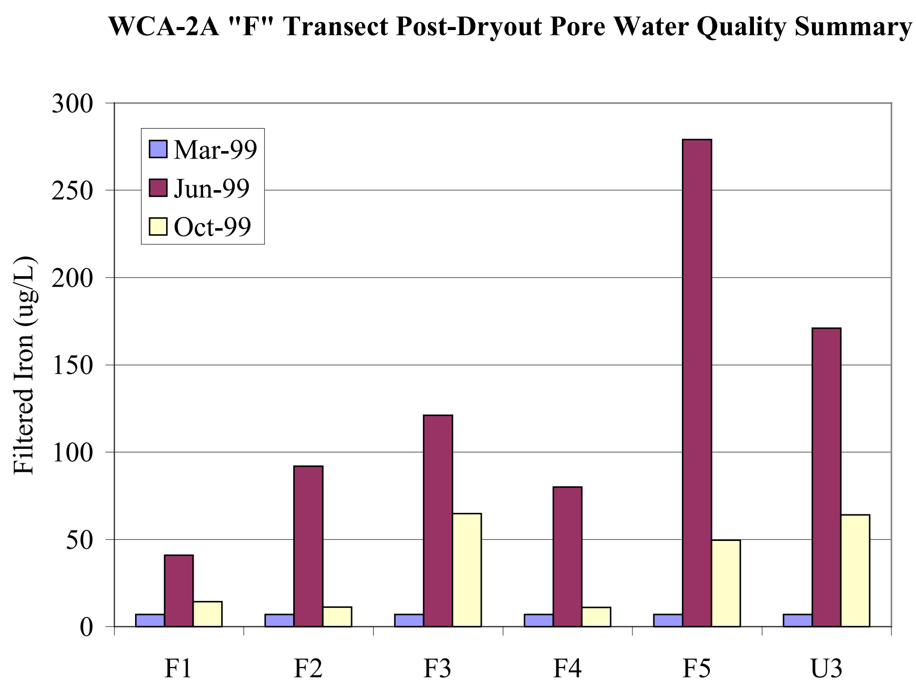

Figure 8. Pore water filtered iron concentrations (10-20 cm) for dryout (March 1999) and post-dryout (March and October 1999) along the "F" transect in WCA-2A

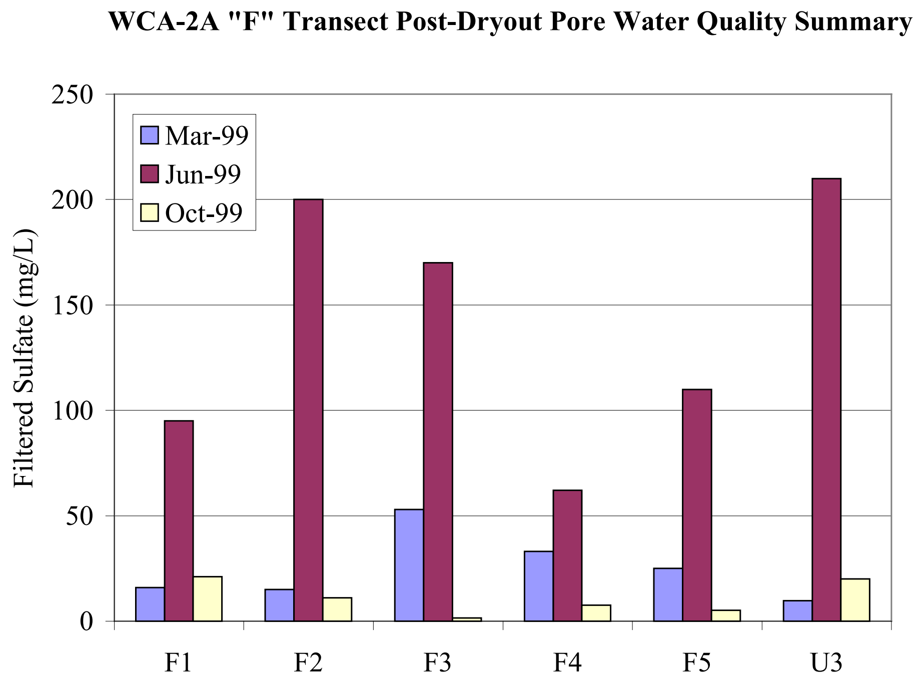

Figure 9. Pore water filtered sulfate concentrations (10-20 cm) for dryout (March 1999) and post-dryout (March and October 1999) along the "F" transect in WCA-2A

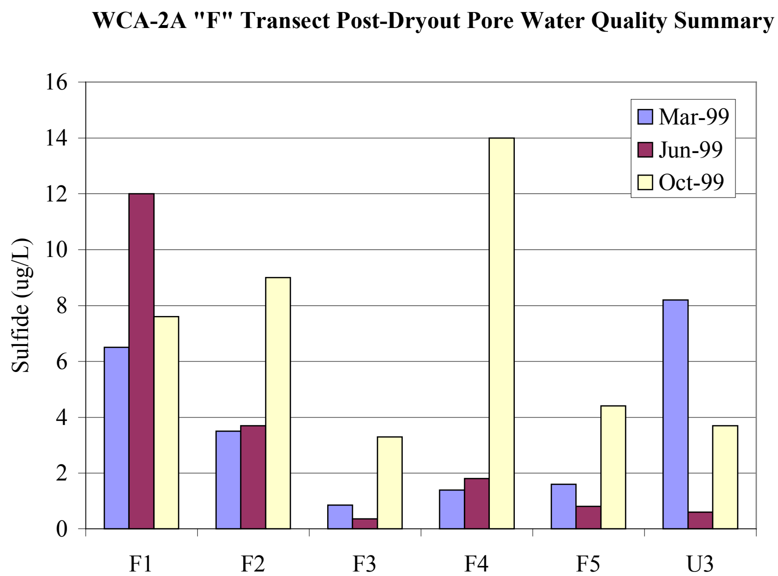

Figure 10. Pore water filtered sulfide concentrations (10-20 cm) for dryout (March 1999) and post-dryout (March and October 1999) along the "F" Transect in WCA-2A

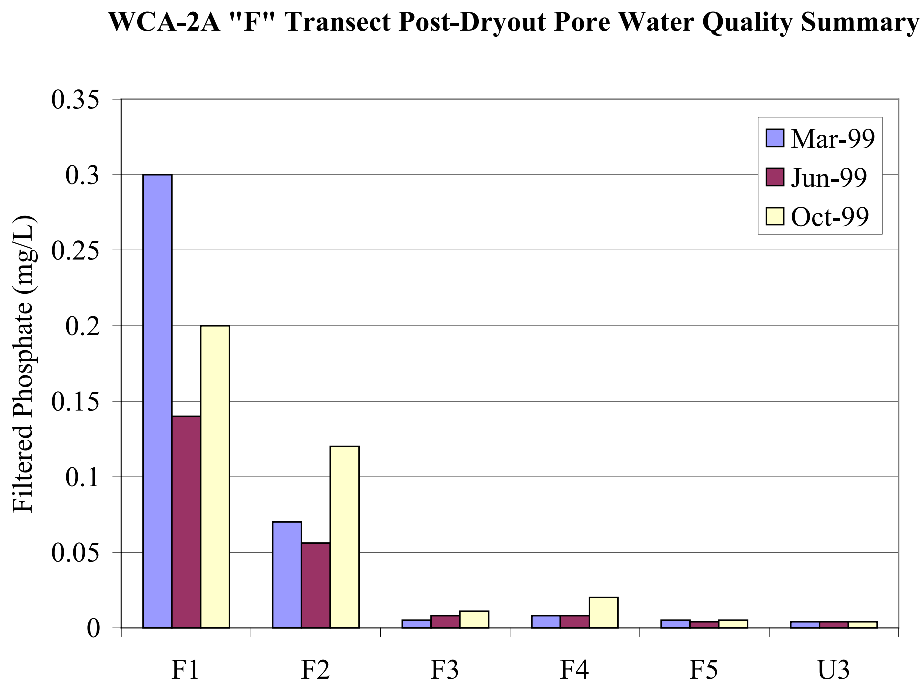

Figure 11. Pore water filtered phosphate concentrations (10-20 cm) for dryout (March 1999) and post-dryout (March and October 1999) along the "F" Transect in WCA-2A

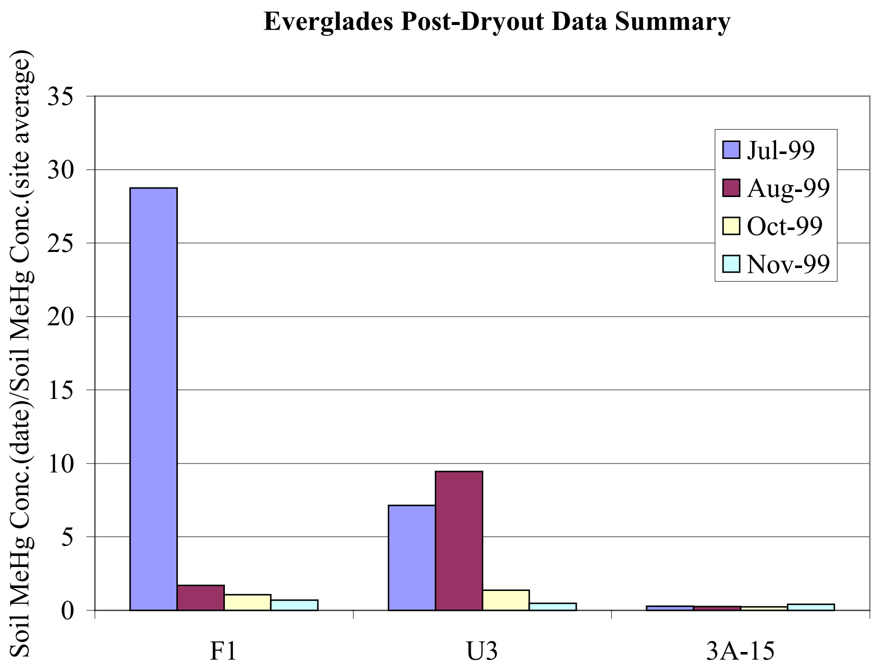

Figure 12. Ratio of post-dryout average concentration of methylmercury (MeHg) in surficial soil to site averages for July-November 1999

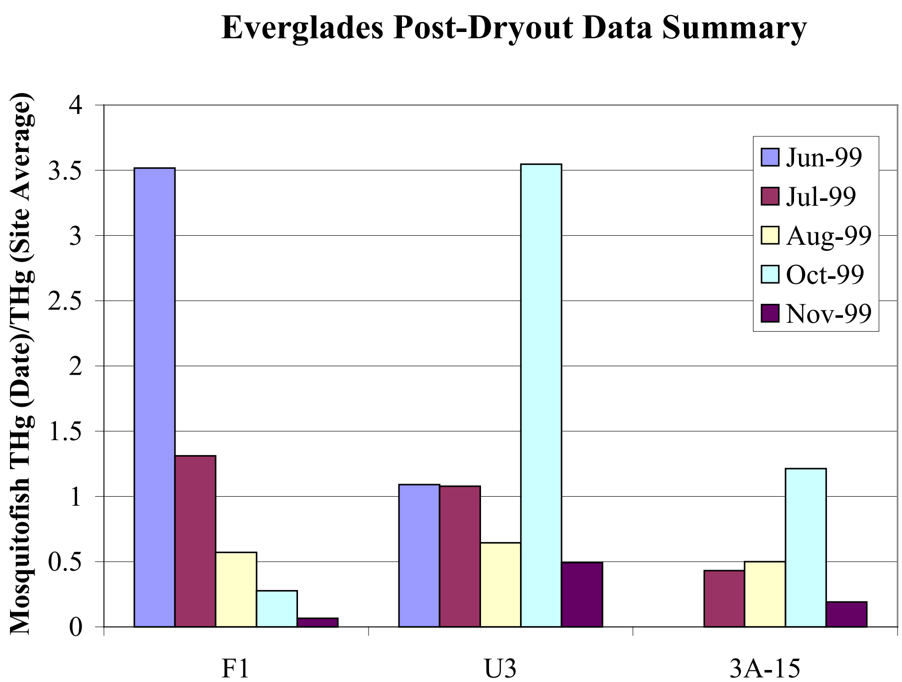

Figure 13. Ratio of post-dryout average concentration of total mercury in mosquitofish to site averages for June-November 1999. [WCA-3A-15 was not routinely monitored by the District in June 1999]

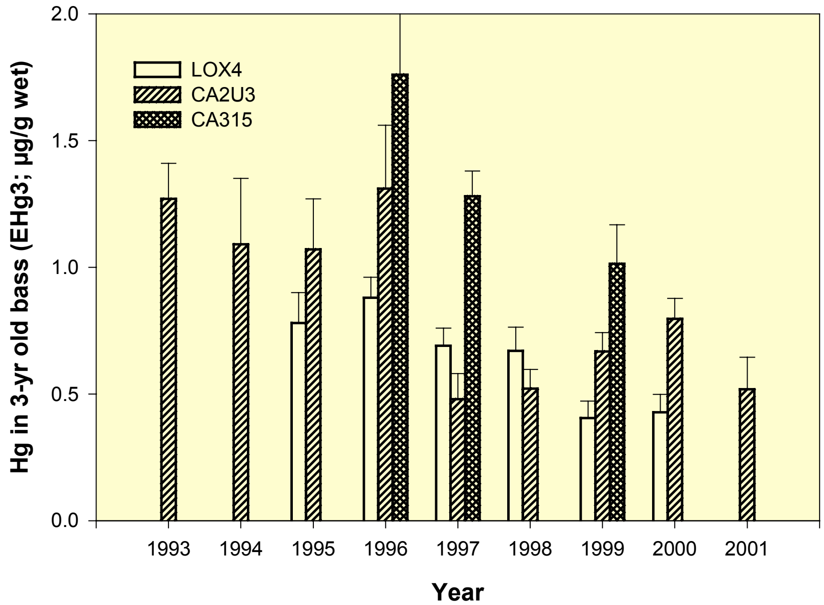

Figure 14. Annual average concentrations of total mercury in largemouth bass at three routinely monitored interior Everglades sites for the period of record

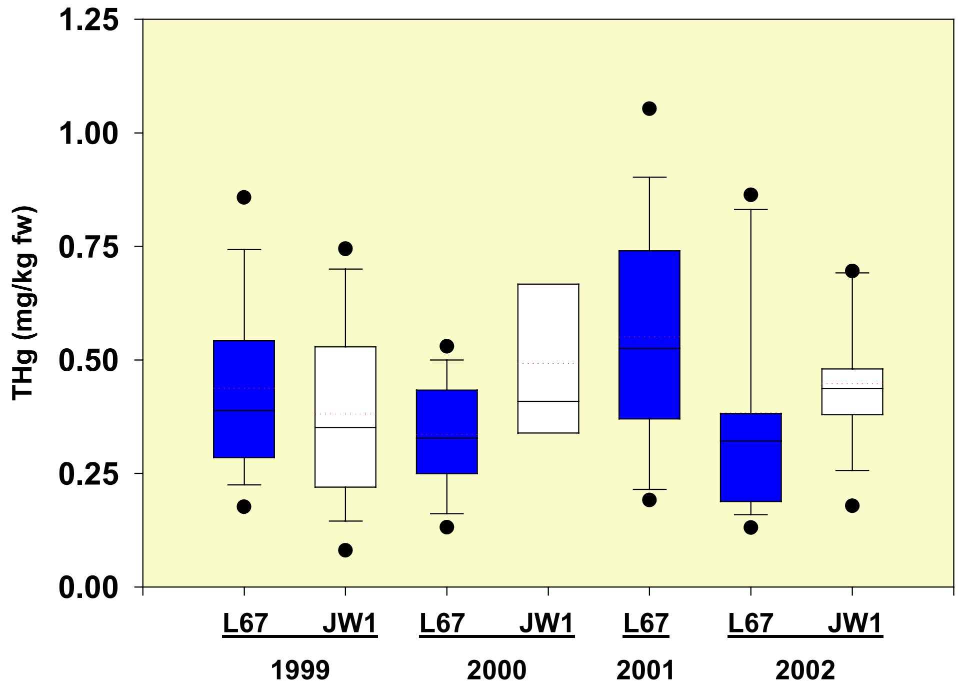

Figure 15. Annual average concentrations of total mercury in great egret eggs at two routinely monitored interior Everglades colonies for the period of record

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